REACTION OF β-ARYLACRYLYLOXIRANES WITH AMIDINES

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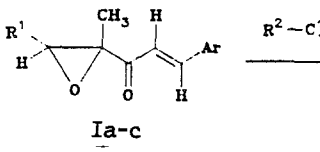
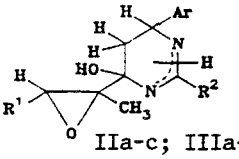
Mixtures of stereoisomers of oxiranylhydroxytetrahydropyrimidines were obtained by cyclocondensation of β-arylacrylyloxiranes with acetamidines or benzamidines; the 2-aryl derivatives of the products give the corresponding dihydropyrimidines and pyrimidines under base-catalysis conditions. In acetic acid dehydration and oxidation are accompanied by opening of the epoxide ring.

The important role of pyrimidine derivatives in biological processes is responsible for the interest of researchers in the chemistry of this class of compounds [1–3]. The aim of the present research was the synthesis of new derivatives of the pyrimidine series that contain an epoxide ring in the side chain, which opens up extensive possibilities for subsequent functionalization.

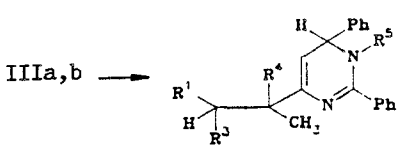
The reaction of β-arylacrylyloxiranes Ia-c with acetamidines or benzamidines leads to mixtures of stereoisomeric 4(6)-hydroxy-2-methyl(phenyl)-6(4)-aryl-4(6)-1,2-epoxyalkyl-1,4,5,6-tetrahydropyrimidines IIa–c and IIIa–c in good yields.



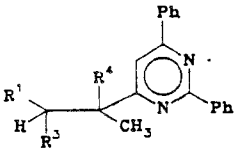
Ia-c



IIa-c; IIIa-c



IVa, b; VIb; VIIa, b; VIIIb; IXa, b



Va,b; Xa

TABLE 1

Compound	R^1	R^3	R^4	R^5	Compound	R^1	R^3	R^4	R^5
IVa	Me	AcO (OH)	OH (AcO)	H	VIIa	Me	OH	OH	Ac
IVb	H	AcO	OH	H	VIIb	H	OH	OH	Ac
Va	Me	AcO (OH)	OH (AcO)	—	VIIIb	H	AcO	AcO	Ac
Vb	H	OH	OH	—	IXa	Me	—O—		H
Vb	H	AcO	AcO	H	IXb	H	—O—		H
					Xa	Me	—O—		—

*I–IIIa R^1 = Me, b, c R^1 = H; a, b Ar = Ph, c Ar = p-BrC_6H_4; IIa–c R^2 = Me, IIIa–c R^2 = Ph.

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TABLE 2. Characteristics of II–X

Compound	Empirical formula	mp, °C	IR spectrum, cm^{-1}	PMR spectrum, δ, ppm (J, Hz)	Yield, %
IIa	$C_{15}H_{20}N_2O_2$	140...141	1640 (C=N), 3370 (NH), 3580 (OH)	1,24 (3H, d, J=6,5, 4^2-CH_3); 1,36 (3H, s, 4^1-CH_3); 1,21 (1H, d.d, J=12,8, J=11,0; 5-H_a); 1,83 (3H, s, 2-CH_3); 1,87 (2H, d.d, J=11,0, J=3,4, 5-H_e); 3,01 (1H, q, J=6,5, 4^2-H); 4,00 (1H, s, OH); 4,35 (1H, d. d J=12,8, J=3,4, 6-H); 5,50 (1H, s, NH); 7,21, 7,31 (5H, m, Ph) 1,17 (3H, d, J=6,5, 4^2-CH_3); 1,20 (3H, s, 4^1-CH_3); 1,22 (1H, d.d., J=11,0, J=12,8, 5-H_a); 1,73 (1H, d. d, J=11,0, J=3,4, 5-H_e); 1,81 (3H, s, 2-CH_3); 3,28 (1H, q, J=6,5, 4^2-H); 4,00 (1H, s, OH); 4,35 (1H, d. d J=12,8; J=3,4, 6-H); 5,50 (1H, s, NH); 7,21, 7,31 (5H, m, Ph)	73
IIb	$C_{14}H_{18}N_2O_2$	146...148	1640 (C=N), 3450 (NH), 3600 (OH)	1,24 (1H, d.d., J=11,0, J=12,8, 5-H_a); 1,31 (3H, s, 4^1-CH_3); 1,83 (3H, s, 2-CH_3); 1,85 (1H, d.d, J=11,0, J=3,4, 5-H_e); 2,45, 2,57 (2H, AB-system J_{AB}=5,0, 4^2-H); 3,37 (1H, s, OH); 4,36 (1H, d. d J=12,8, J=3,4, 6-H); 5,41 (1H, s, NH); 7,19, 7,31 (5H, m, Ph) 1,24 (1H, d. d, J=11,0, J=12,8, 5-H_a); 1,35 (3H, s, 4^1-CH_3); 1,70 (3H, s, 2-CH_3); 1,73 (1H, d.d, J=11,0, J=3,4, 5-H_e); 2,45, 2,95 (2H, AB-system, J_{AB}=5,0; 4^2-H); 3,37 (1H, s, OH); 4,36 (1H, d.d, J=12,8, J=3,4, 6-H); 5,41 (1H, s, NH); 7,19, 7,31 (5H, m, Ph)	77
IIc	$C_{14}H_{17}BrN_2O_2$	167...168	1640 (C=N), 3440 (NH), 3580 (OH)	1,24 (1H, d.d., J=11,0, J=12,8, 5-H); 1,31 (3H, s, 4^1-CH_3); 1,85 (1H, d. d. J=11,0, J=3,4, 5-H_e); 1,80 (3H, s, 2-CH_3); 2,43, 2,71 (2H, AB-system, J_{AB}=5,0; 4^2-H); 3,37 (1H s, OH); 4,36 (1H, d.d, J=12,8, J=3,4, 6-H); 5,41 (1H, s, NH); 7,31 (4H, m, Ph) 1,24 (1H, d. d J=11,0, J=12,8, 5-H_a); 1,35 (3H, s, 4^1-CH_3); 1,70 (3H, s, 2-CH_3); 1,73 (1H, d.d, J=11,0, J=3,4, 5-H_e); 2,43, 2,93 (2H, AB-system, J_{AB}=5,0; 4^2-H); 3,37 (1H, s, OH); 4,36 (1H, d.d, J=12,8, J=3,4, 6-H); 5,41 (1H, s, NH); 7,31 (4H, m, Ph)	60
IIIa	$C_{20}H_{22}N_2O_2$	124...126	1630 (C=N), 3360 (NH), 3420 (OH)	1,18 (3H, d, J=6,5 4^2-CH_3); 1,16 (1H, d.d. J=12,0, J=12,5, 5-H_a); 1,36 (3H, s, 4-CH_3); 1,78 (1H, d.d. J=12,0, J=4,0, 5-H_e); 2,98 (1H, q, J=6,5, 4^2-H); 4,60 (1H, d.d. J=12,5, J=4,0, 6-H); 6,00 (1H, s NH); 7,20, 7,78 (10H, m, Ph) 1,18 (3H, d. J=6,5, 4^2-CH_3); 1,16 (1H, d.d. J=12,0, J=12,5, 5-H_a); 1,36 (3H, s 4-CH_3); 2,12 (1H, d.d., J=12,0, J=4,0, 5-H); 3,30 (1H, q J=6,5, 4^2-H); 4,60 (1H, d.d., J=12,5, J=4,0, 6-H); 6,00 (1H, s, NH); 7,20, 7,78 (10H, m, Ph)	98
IIIb	$C_{19}H_{20}N_2O_2$	130...133	1630 (C=N), 3370 (NH), 3450 (OH)	1,40 (3H, s, 4^1-CH_3); 1,45 (1H, t, J=12,0, 5-H_a); 2,03 (1H, m, 5-H_e); 2,47, 2,58 (2H, AB-system, J_{AB}=5,0, 4^2-H); 4,64 (1H, d.d. J=4,0, J=12,0, 6-H); 5,50 (1H. s, NH); 7,34, 7,80 (10H, m, Ph) 1,38 (3H, s, 4^1-CH_3); 1,45 (1H, t, J=12,0, 5-H_a); 1,90 (1H, d d. J=4,0, J=12,0, 5-H_e); 2,40, 3,04 (2H, AB-system, J_{AB}=5,0, 4^2=H); 4,64 (1H, d.d. J=4,0, J=12,0, 6-H); 5,50 (1H, s, NH); 7,30, 7,80 (10H, m, Ph)	76

TABLE 2. (cont'd)

Compound	Empirical formula	mp, °C	IR spectrum, cm^{-1}	PMR spectrum, δ, ppm (J, Hz)	Yield, %
IIIc	$C_{19}H_{19}BrN_2O_2$	142...147	1630 (C=N), 3370 (NH), 3450 (OH)	1,36 (3H, s, 4^1-CH_3); 1,36 (1H, m, 5-H_a); 1,98 (1H, m, 5-H_e); 2,40, 2,70 (2H, AB-system, J_{AB}=5,0, 4^2-H); 4,60 (1H, d. d. J=4,0, J=12,0, 6-H); 7,30, 7,80 (11H, m, Ph, NH, OH) 1,34 (3H, s, 4^1-CH_3); 1,36 (1H, m, 5-H_a); 1,98 (1H, m, 5-H_e); 2,40, 3,02 (2H, AB-system, J_{AB}=5,0, 4^2-H); 4,60 (1H, d.d., J=4,0, J=12,0, 6-H); 7,30, 7,80 (11H, m, Ph, NH, OH)	59
IVa	$C_{22}H_{24}N_2O_3$	Oil	1740 (C=O), 3380 (NH), 3440 (OH)	1,23 (3H, d, J=6,5, 4^2-CH_3); 1,32 (3H, s, 4^1-CH_3); 2,02 (3H, s, CH_3CO); 4,92, 5,29 (2H, 2 d, J=3,6, 5-, 6-H); 5,04 (1H, q, J=6,5; 4^2-H); 5,46 (1H, s, OH); 7,50 (11H, m, Ph, NH) 1,23 (3H, d, J=6,5, 4^2-CH_3); 1,32 (3H, s, 4^1-CH_3); 1,72 (3H, s, CH_3CO); 4,93 (1H, q, J=6,5, 4^2-H); 4,37, 5,29 (2H, 2 d, J=6,5, 5-, 6-H); 5,46 (1H, m, OH); 7,50 (11H, m, Ph, NH)	60
IVb	$C_{21}H_{22}N_2O_3$	Oil	1740 (C=O), 3380 (NH), 3420 (OH)	1,34 (3H, s, 4^1-CH_3); 1,84 (3H, s, CH_3CO); 4,00, 4,14 (2H, AB-system, J_{AB}=11,5, 4^2-H); 5,08, 5,30 (2H, 2 d, J=4,0, 5-, 6-H); 5,52 (1H, s, OH); 7,20, 7,80 (11H, m, Ph, NH)	77
Va	$C_{22}H_{22}N_2O_3$	120...122	1740 (C=O), 1620 (N=C), 3420 (OH)	1,06 (3H, d, J=6,6, 4^2-CH_3); 1,60 (3H, s, 4^1-CH_3); 1,98 (3H, s, CH_3CO); 3,80 (1H, s, OH); 5,40 (1H, q, J=6,6, 4^2-H); 7,40, 8,20, 8,50 (10H, m, Ph); 8,06 (1H, s, 5-H)	25
Vb	$C_{21}H_{20}N_2O_3$	78...80	1730 (C=O), 1620 (N=C), 3420 (OH)	1,60 (3H, s, 4^1-CH_3); 1,84 (3H, s, CH_3CO); 4,38, 4,45 (2H, AB-system, J_{AB}=12,0, 4^2H); 5,20 (1H, s, OH); 7,40, 8,20, 8,50 (10H, m, Ph); 8,06 (1H, s, 5-H)	94
VIb	$C_{23}H_{24}N_2O_4$	143...145	1740 (C=O), 3370 (NH)	1,62 (3H, s, 4^1-CH_3); 1,90 (3H, s, CH_3CO); 1,92 (3H, s, CH_3CO); 4,42 (2H, s, 4^2-H); 5,03, 5,25 (2H, 2 d, J=4,0, 5-, 6-H); 7,26, 7,75 (11H, m, Ph, NH)	51
VIIa	$C_{24}H_{26}N_2O_4$	158...160	1680 (C=O), 1740 (C=O), 3480 (OH)	1,30 (3H, d, J=6,5, 4^2-CH_3); 1,54 (3H, s, 4^1-CH_3); 1,70 (3H, s, CH_3CO); 1,80 (3H, s, CH_3CO); 5,33 (1H, q, J=6,5, 4^2-H); 6,00, 6,30 (2H, 2 d, J=6,5, 5-, 6-H); 7,30, 7,35 (10H, Ph)	47
VIIb	$C_{23}H_{24}N_2O_4$	139...140	1680 (C=O), 1740 (C=O), 3480 (OH)	1,56 (3H, s, 4^1-CH_3); 1,70 (3H, s CH_3CO); 1,92 (3H, s CH_3CO); 4,04, 4,46 (2H, AB-system J_{AB}=10,8, 4^2-H); 4,20 (1H, s, OH); 6,00, 6,30 (2H, 2 d, J=6,5, 5-, 6-H); 7,32 (10H, m, Ph)	44
VIIIb	$C_{25}H_{25}N_2O_5$	158...160	1685 (C=O), 1740 (C=O), 3480 (OH)	1,76 (3H, s, 4^1-CH_3); 1,86 (3H, s CH_3CO); 2,05 (3H, s, CH_3CO); 2,13 (3H, s, CH_3CO); 4,55, 4,71 (2H, AB-system J=11,5); 5,82, 6,39 (2H, 2 d, J=6,5, 5-, 6-H); 7,32 (10H, m, Ph)	42
IXa	$C_{20}H_{20}N_2O$	Oil	1680 (C=N), 3430 (NH)	1,15 (3H, d, J=6,6, 4^2-CH_3); 1,36 (3H, s, 4^1-CH_3); 2,86 (1H, q J=6,6, 4^2-H); 4,90, 5,16 (2H, AB-system, J_{AB}=4,0, 5-, 6-H); 6,30 (1H, s, NH); 7,20 (10H, m, Ph)	65
IXb	$C_{19}H_{18}N_2O$	Oil	1680 (C=N), 3430 (NH)	1,42 (3H, s, 4^1-CH_3); 2,52, 2,86 (2H, AB-system, J_{AB}=5,0); 5,00, 5,20 (2H, AB-system, J_{AB}=4,0, 5-, 6-H); 6,05 (1H, s, NH); 7,20 (10H, m, Ph)	75
Xa	$C_{20}H_{18}N_2O$	93...94	1630 (C=N)	1,40 (2H, d, J=6,5, 4^2-CH_3); 1,75 (3H, s, 4^1-CH_3); 3,08 (1H, q, J=6,5, 4^2-H); 7,35, 8,28, 8,58 (10H, m, Ph); 7,55 (1H, s, 5-H)	89

The absorption bands of the enone system that are characteristic for epoxy ketones Ia–c vanish in the IR spectra of IIa–c and IIIa–c, and absorption bands of C=N (1640 cm^{-1}) and NH and OH (3360–3450 and 3450–3600 cm^{-1}) groups appear (Table 2). The PMR spectra of oxiranyltetrahydropyrimidines IIa–c and IIIa–c contain an ABX system of signals of protons of a pyrimidine ring, which show up in the form of three groups of signals, the chemical shifts and spin-spin coupling constants (SSCC) of which correspond to the literature data [4]. The chemical shifts and SSCC of the protons of the epoxide rings of IIa–c and IIIa–c constitute evidence for retention of the oxirane ring during the reaction.

The dehydration of oxiranylhydroxytetrahydropyrimidines IIa–c and IIIa–c to the corresponding dihydropyrimidines with retention of the epoxide ring could not be accomplished using the methods described in the literature, viz., by heating in benzene or toluene with removal of water by azeotropic distillation [5] or by refluxing in an aprotic solvent with 4-Å molecular sieves [6]. On heating in acetic acid [6], the dehydration of IIIa, b is accompanied by opening of the epoxide ring with the formation of β-acetoxy-α-hydroxyalkyldihydropyrimidine IVb or a mixture of β-hydroxy-α-acetoxy- and β-acetoxy-α-hydroxyalkyldihydropyrimidines IVa. In the PMR spectra of IVa, b the signals of the 5-H and 6-H protons of the dihydropyrimidine ring show up in the form of an AB system with SSCC 3.5–4.0 Hz at 4.90–5.30 ppm, a range that, according to the data in [5, 7], is characteristic for one tautomeric form of the pyrimidine ring. When they are allowed to stand in air in an aprotic solvent, IVa, b are readily oxidized to the corresponding pyrimidines Va, b, in the PMR spectra of which, instead of the AB system of signals of protons of the heterocyclic ring, a singlet signal of the 5-H proton of the pyrimidine ring appears at 8.06 ppm. Tetrahydropyrimidine IIIb is converted to 1,2-diacetoxyisopropyldihydropyrimidine VIb by the action of acetic acid–acetic anhydride.

When hydroxytetrahydropyrimidines IIIa, b are refluxed in acetic anhydride, N-acetylation of the dihydropyrimidine ring with the liberation of monoacetoxy-N-acetylpyrimidines VIIa, b occurs in addition to dehydration and opening of the epoxide ring; an increase in the reaction time to 12 h in the case of IIIb leads to the formation of a mixture of mono- and diacetoxy-N-acetylpyrimidines VIIb and VIIIb. The change in the chemical shifts of the 5-H and 6-H protons in the PMR spectra of N-acetyldihydropyrimidines VIIa, b as compared with dihydropyrimidines IVa, b indicates localization of the N-acetyl group in the dihydropyrimidine ring.

We were able to accomplish the synthesis of oxiranyldihydropyrimidines IXa, b by the reaction of hydroxytetrahydropyrimidines IIIa, b with tetrabutylammonium hydroxide in dioxane; the subsequent aromatization of the dihydropyrimidines proceeds under the influence of oxygen. Thus, IXa is oxidized to oxiranylpyrimidine Xa in acetone in air after 7 days.

EXPERIMENTAL

The IR spectra of solutions of the compounds in CCl_4 and $CHCl_3$ ($c = 10^{-1}$ mole/liter, $l = 0.01$ cm) were recorded with a Specord IR-75 spectrometer. The NMR spectra of solutions of the compounds in $CDCl_3$, d_6-acetone, and d_6-DMSO were obtained with Bruker WM-360 (360 MHz) and Tesla BS-467A (60 MHz) spectrometers with tetramethylsilane (TMS) as the internal standard. The mass spectra of IIb and IIIb were recorded with a Varian MAT-311 spectrometer at an ionizing voltage of 70 eV.

4(6)-Hydroxy-2-methyl-6(4)-aryl-4(6)-1,2-epoxyalkyl-1,4,5,6-tetrahydropyrimidines IIa–c and 4(6)-Hydroxy-2-phenyl-6(4)-aryl-4(6)-1,2-epoxyalkyl-1,4,5,6-tetrahydropyrimidines IIIa–c. A 0.02-mole sample of epoxyenone Ia–c was dissolved in 30 ml of absolute acetone, and the solution was added dropwise at 0–10°C with constant stirring to 0.025 mole of the acetamidine or benzimidine in 25 ml of absolute acetone. After 2–3 h, the precipitated IIa–c or IIIa–c was removed by filtration and washed with ether. A second portion of crystals was isolated by partial evaporation of the solution. Compounds IIa–c and IIIa–c were recrystallized from acetone.

^{13}C NMR spectrum (mixture of stereoisomers): 12.67, 13.12, 13.63, 13.75 (4 q, 4^1-, 4^2-CH_3); 22.17 (q, 2-CH_3); 38.14, 39.05 (2t, $C_{(5)}$); 51.47, 52.20, 53.59, 54.20 (4d, $C_{(6)}$, $C_{(42)}$); 63.69, 63.79 (2s, $C_{(41)}$); 78.68, 79.17 (2s, $C_{(4)}$); 126.09, 126.67, 127.92, 128.08 (6d, o-, m-, p-Ph); 144.77, 145.64 (2s, i-Ph); 153.03, 153.98 (2s, $C_{(2)}$). ^{13}C NMR spectrum of IIIb: 17.16 (q, 4^1-CH_3); 22.26 (q, 2-CH_3); 37.93 (t, $C_{(5)}$); 50.30 (t, $C_{(42)}$); 52.34 (d, $C_{(6)}$); 60.54 (s $C_{(41)}$) 78.15 (s, $C_{(4)}$); 126.48, 126.73, 127.99 (3d, o-, m-, p-Ph); 145.70 (s, i-Ph); 152.99 (s, $C_{(2)}$).

2,6-Diphenyl-4-(2(1)-acetoxy-1(2)-hydroxyalkyl)-1,6-dihydropyrimidines IVa, b. A 0.01-mole sample of IIIa, b was heated for 3–5 h at 80°C in 5 ml of glacial acetic acid, after which the mixture was diluted with a tenfold amount of water, and the aqueous mixture was neutralized with sodium carbonate solution. The mixture was then extracted with ether (three 50-ml portions), and the extract was dried over sodium sulfate. After evaporation of the ether to 25–30 ml, the residue was filtered through a layer of 100/250 μm silica gel (25 × 50 mm) by elution with ether–hexane (1:1).

Compounds IVa, b were isolated in the form of oil products after evaporation of the solvent. Dihydropyrimidines IVa were a mixture of 4-(2-acetoxy-1-hydroxy-1-methylpropyl)- and 4-(1-acetoxy-2-hydroxy-1-methylpropyl)-2,6-diphenyl-1,6-dihydropyrimidine in a ratio of 2:1.

2,6-Diphenyl-4-(2-acetoxy-1-hydroxyalkyl)pyrimidines (Va, b). A 0.005-mole sample of dihydropyrimidines IVa, b was maintained in 2 ml of acetone for 2 days, after which the acetone was evaporated, and pyrimidines Va, b were crystallized from methyl ethyl ketone–ether.

2,6-Diphenyl-4-(1,2-diacetoxyisopropyl)-1,6-dihydropyrimidine (VIb). A 0.01-mole sample of IIIb was dissolved in a mixture of 5 ml of acetic acid and 5 ml of acetic anhydride, and the solution was allowed to stand for 12 h. It was then diluted with water to 100 ml, and the aqueous mixture was neutralized with sodium carbonate and extracted with ether (three 50-ml portions). The ether solution was washed with water and dried over sodium sulfate. After evaporation of the ether to 20 ml, 20 ml of hexane was added, and VIb was removed by filtration.

1-Acetyl-2,6-diphenyl-4-(2-acetoxy-1-hydroxyalkyl)-1,6-dihydropyrimidines (VIIa, b). A 7-ml sample of acetic anhydride was added to 0.01 mole of tetrahydropyrimidine IIIa, b, and the mixture was refluxed for 5 h, after which it was worked up as described above. Compound VIIa was isolated by means of chromatography with a column (2 × 10 cm) packed with 100/250 μm silica gel by elution with ether and had R_f 0.31; VIIb had R_f 0.08.

1-Acetyl-2,6-diphenyl-4-(1,2-diacetoxyisopropyl)-1,6-dihydropyrimidine (VIIIb). A 0.01-mole sample of tetrahydropyrimidine IIIb was refluxed for 12 h in 7 ml of acetic anhydride, after which the reaction mixture was worked up as described above. Compound VIIIb was isolated by means of column chromatography (R_f 0.21) in the same way as VIIb and was recrystallized from ether–hexane (2:1).

2,6-Diphenyl-4-(1,2-epoxyalkyl)-1,6-dihydropyrimidines (IXa, b). A 0.2–0.3 mole sample of tetrabutylammonium hydroxide was added to 0.01 mole of tetrahydropyrimidine IIIa, b, and the mixture was heated until the solid material had dissolved. After 12 h, the dioxane was removed, and the residue was filtered through a layer of silica gel (2.5 × 3 cm) by elution with ether. The ether was evaporated, and IXa, b were isolated in the form of yellow oily products with $R_{f\,IXa}$ 0.37 and $R_{f\,IXb}$ 0.35 [Silufol, acetone–ether (1:1)].

2,6-Diphenyl-4-(1,2-epoxyalkyl)pyrimidines (Xa, b). These compounds were obtained by maintaining 5 mmoles of IXa, b in 5 ml of acetone in air for 7 days. Evaporation of the acetone gave pyrimidines Xa, b, which were crystallized from methyl ethyl ketone–ether (1:4).

LITERATURE CITED

1. S. K. Robev, *Khim. Geterotsikl. Soedin.*, No. 12, 1587 (1981).
2. H. Cho, K. Shima, Y. Ohnaka, A. Mizuno, and Y. Takeuchi, *J. Org. Chem.* **50**, 4227 (1985).
3. H. Cho, M. Ueda, K. Shima, A. Misuno, M. Hayashimatsu, Y. Ohnaka, and Y. Takeuchi, *Heterocycles* **24**, 233 (1986).
4. A. L. Weis and V. P. Mamaev, *Khim. Geterotsikl. Soedin.*, No. 5, 674 (1977).
5. A. L. Weis, V. M. Shirina, and V. P. Mamaeva, *Izv. Sibirsk. Otd. Akad. Nauk SSSR, Ser. Khim. Nauk*, No. 14, 144 (1975).
6. A. L. Weis, *Synthesis*, No. 5, 528 (1985).
7. A. L. Weis, Z. Porat, and Z. Luz, *J. Am. Chem. Soc.* **106**, 8021 (1984).